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Absence of Crystalline Order in Two-Dimensions

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Abstract

It is proved that in two dimensions a system of electrons embedded in a uniform neutralizing positive background and interacting by a potential given by e^2/r cannot exhibit long range crystalline order at any finite temperature.

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Grimes and Adams[1] have recently presented evidence for a crystalline transition of electrons trapped on a surface of liquid helium. This quasi two-dimensional system has been canonically modelled as a system of electrons in two dimensions, interacting by a potential given by e^2/r, and neutralized by a uniform positive background. Here r is the two dimensional distance between two points. In this context it is important to know if at any finite temperature such a system can display true long-range crystalline order in the thermodynamic limit.

We want to emphasize that the crystalline order in this system is not ruled out by the classic work of Mermin[2]. However, from arguments due to Landau[3] and Peierls[4] it is simple to show that a harmonic solid cannot exist in the thermodynamic limit. Although the arguments by Landau and Peierls can be challenged as not being rigorous enough, a recent Monte Carlo simulation by Gann, Chakravarty and Chester[5] indicated, although not conclusively, the validity of the conclusion drawn from the Landau-Peierls argument. On the basis of the numerical work it was conjectured in ref. 5 that although a rigorous proof does not exist, the general conclusion of Landau and Peierls should be valid.

In this paper we shall prove rigorously that a true long-range crystalline order cannot exist in the thermodynamic limit, thus placing Landau-Peierls argument on a firmer basis for the interparticle interaction mentioned above. The proof makes use of the Bogoliubov's inequality[6] as discussed by Mermin[2]. Although the proof differs in some essential aspects from that of Mermin, the general strategy is similar. We therefore follow him quite closely, making essential changes where necessary.

Because of the long-range nature of the interaction, some complications arise. Although these complications are well known, it is important to state them clearly. (a) In order to obtain physically meaningful results the interaction e^2/r is replaced by $\frac{e^2}{r}\exp(-\mu r)$. Since we are interested in the bulk properties

of the neutral medium our limiting procedure will be first $N \to \infty$, $A \to \infty$, $N/A \to n$ = constant and then $\mu \to 0$. Here N is the total number of electrons and A the area. (b) Another related assumption[7] is that the equilibrium state of the system does not have macroscopic surface charge. Although it is highly implausible that an equilibrium state violating this assumption could have lower free energy, we are not aware of a proof of this.

It will become evident later that even with these assumptions stated above, the original proof of Mermin does not go through because of the plasmons in the long wavelength limit. Consider, then, N classical electrons enclosed in a box of area A. A uniform neutralizing positive background is assumed to exist. The interaction energy U is,

$$U = \frac{1}{2} \sum_{i \neq j} \frac{e^2}{|\underset{\sim}{r}_i - \underset{\sim}{r}_j|} \exp(-\mu|\underset{\sim}{r}_i - \underset{\sim}{r}_j|) - \frac{N}{A} \sum_{i=1}^{N} \int_A d\underset{\sim}{r} \frac{e^2}{|\underset{\sim}{r} - \underset{\sim}{r}_i|} \exp(-\mu|\underset{\sim}{r} - \underset{\sim}{r}_i|)$$

$$+ \frac{N^2}{2A^2} \int_A d\underset{\sim}{r} \int_A d\underset{\sim}{r}' \frac{e^2}{|\underset{\sim}{r} - \underset{\sim}{r}'|} \exp(-\mu|\underset{\sim}{r} - \underset{\sim}{r}'|) \tag{1}$$

In Eq. (1) the first term is the electron-electron interaction, the second the interaction between the electrons and the positive background, and the third the self energy of the background.

We adopt the same criterion for crystallinity as Mermin, i.e.,

$$\lim_{N \to \infty} \frac{\langle \rho_k \rangle}{N} = 0, \ \underset{\sim}{k} \text{ not a reciprocal lattice vector,}$$

$$\lim_{N \to \infty} \frac{\langle \rho_k \rangle}{N} \neq 0, \text{ for at least one reciprocal lattice vector } \underset{\sim}{G},$$

where,

$$\rho_{\underset{\sim}{k}} = \sum_{i=1}^{N} e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}_i} \tag{2}$$

Here $\langle \rangle$ denotes the canonical ensemble average with respect to the interaction energy U and the integrations are over the interior of a box of area A. We now consider the Schwartz inequality

$$\langle |C|^2 \rangle \geq \frac{\langle |C^* \underset{\sim}{B}|^2 \rangle}{\langle |\underset{\sim}{B}|^2 \rangle} \tag{3}$$

and choose C and $\underset{\sim}{B}$ to be,

$$C = \sum_{i=1}^{N} e^{-i(\underset{\sim}{k}+\underset{\sim}{G})\cdot\underset{\sim}{r}_i} \tag{4}$$

$$\underset{\sim}{B} = -k_B T e^{\beta U} \sum_{i=1}^{N} \left(\nabla_i - \frac{\underset{\sim}{k}(\underset{\sim}{k}\cdot\nabla_i)}{k^2}\right)\left(f(\underset{\sim}{r}_i) e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}_i} e^{-\beta U}\right) \tag{5}$$

The function $f(\underset{\sim}{r}_i)$ is chosen as follows[7]. Consider Ω_a to be a set of points within a distance $aL^{\frac{1}{2}}$ of the walls of the box, where a is independent of L. f is now defined by the following conditions: (1) $0 \leq f \leq 1$ in Ω_a, (2) $f = 1$ everywhere else, and (3) $|\nabla f| < bL^{-\frac{1}{2}}$ in Ω_a, for some b independent of L. It will become clearer later on that we have gone into great pains in introducing the functions $f(\underset{\sim}{r}_i)$. The intention has been to make the surface terms arising from the integrations by parts vanish. The complications arising from $f(\underset{\sim}{r}_i)$ could have been entirely avoided with a slightly careless use of the periodic boundary condition. The choice of $\underset{\sim}{B}$, Eq. (5), is one of the key points of our proof and serves to project out the longitudinal part of the force and suppresses the plasmons in the denominator of the right hand side of the inequality, Eq. (3). With these choices of C and $\underset{\sim}{B}$, Eqs. (4) and (5), it is straightforward to derive the inequality

$$\frac{1}{N} \langle |\rho_{\underset{\sim}{k}+\underset{\sim}{G}}|^2 \rangle \geq k_B T \text{ [illegible]} \tag{6}$$

where

$$\Delta(\underline{G}) = [G^2-(\hat{k}\cdot\underline{G})^2]\left|\frac{1}{N}\int d\underline{r}_1\, P(\underline{r}_1)\, f(\underline{r}_1)\, e^{i\underline{G}\cdot\underline{r}_1}\right|^2 \quad . \tag{7}$$

$$\Lambda(\underline{k}) = \frac{k_B T}{N}\int d\underline{r}_1\, P(\underline{r}_1)[(\nabla_1\, f(\underline{r}_1))^2 - (\hat{k}\cdot\nabla_1\, f(\underline{r}_1))^2]$$

$$-\frac{n}{N}\int d\underline{r}_1\, f^2(\underline{r}_1)\, P(\underline{r}_1)\, [\nabla_1^2 - (\hat{k}\cdot\nabla_1)^2]\int d\underline{r}_2\, \frac{e^{-\mu r_{12}}}{r_{12}}$$

$$+\frac{1}{2N}\int d\underline{r}_1\int d\underline{r}_2\, |f(\underline{r}_1)e^{i\underline{k}\cdot\underline{r}_1} - f(\underline{r}_2)e^{i\underline{k}\cdot\underline{r}_2}|^2\, P(\underline{r}_1,\underline{r}_2)[\nabla_1^2-(\hat{k}\cdot\nabla_1)^2]\frac{e^{-\mu r_{12}}}{r_{12}} \tag{8}$$

The distribution functions are defined to be,

$$P(\underline{r}_1) = N\,\frac{\int e^{-\beta U}d\underline{r}_2 \ldots d\underline{r}_N}{\int e^{-\beta U}d\underline{r}_1 \ldots d\underline{r}_N} \quad . \tag{9}$$

$$P(\underline{r}_1,\underline{r}_2) = N(N-1)\,\frac{\int e^{-\beta U}d\underline{r}_3 \ldots d\underline{r}_N}{\int e^{-\beta U}d\underline{r}_1 \ldots d\underline{r}_N} \tag{10}$$

The inequality (6) is now multiplied by a positive Gaussian $g(\underline{k}+\underline{G})$ centered at $\underline{k}+\underline{G}=0$, and summed over $\underline{k}$ to get

$$\frac{1}{A}\sum_{\underline{k}} g(\underline{k}+\underline{G})\,\frac{\langle|\rho_{\underline{k}+\underline{G}}|^2\rangle}{N} \geq \frac{k_B T g(\tfrac{1}{2}G)}{A}\sum_{k<\frac{1}{2}G}\frac{\Delta(\underline{G})}{\Lambda(\underline{k})} \tag{11}$$

The strategy now will be to proceed to the thermodynamic limit, show that $\Lambda(\underline{k}) \sim \lambda k^2$ for small k and then take the limit $\mu \to 0$. The proof will then be completed by showing that the left hand side of the inequality is bounded. Proceeding to the thermodynamic limit then allows us to write

$$\int_0^\infty k\,dk\int_0^{2\pi}d\theta_k\, g(\underline{k}+\underline{G})\,\frac{\langle|\rho_{\underline{k}+\underline{G}}|^2\rangle}{N} \geq k_B T g(\tfrac{1}{2}G)\int_{k<G/2}k\,dk\int_0^{2\pi}d\theta_k\,\frac{\Delta(\underline{G})}{\Lambda(\underline{k})} \tag{12}$$

The inequality (12) can be strengthened further with the help of tricks similar to those used by Fernandez[8] and obtain

$$\int d\underline{k}\, g(\underline{k}+\underline{G})\,\frac{\langle|\rho_{\underline{k}+\underline{G}}|^2\rangle}{N} \geq \frac{\pi k_B T G^2}{8}\, g(\tfrac{1}{2}G)\left(\frac{1}{N}\int d\underline{r}_1 P(\underline{r}_1) f(\underline{r}_1) e^{i\underline{G}\cdot\underline{r}_1}\right)^2$$

$$\int_{k<G/2}k\,dk\,\frac{1}{\left(\frac{4}{\pi}\int_0^{2\pi}d\theta_k\,\Lambda(\underline{k})\right)} \tag{13}$$

Now,

$$\lim_{k\to 0}\ \frac{4}{\pi}\int_0^{2\pi}d\theta_k\,\Lambda(\underline{k}) = \frac{4k_B T}{N}\int d\underline{r}_1 P(\underline{r}_1)(\nabla_1 f(\underline{r}_1))^2 - \frac{4n}{N}\int d\underline{r}_1 f^2(\underline{r}_1)P(\underline{r}_1)\nabla_1^2\int d\underline{r}_2\,\frac{e^{-\mu r_{12}}}{r_{12}}$$

$$+\frac{2}{N}\int d\underline{r}_1\int d\underline{r}_2\,\frac{e^{-\mu r_{12}}}{r_{12}}(\mu^2 r_{12}^2+\mu r_{12}+1)[f(\underline{r}_1)-f(\underline{r}_2)]^2 P(\underline{r}_1,\underline{r}_2)$$

$$+\frac{k^2}{2N}\int d\underline{r}_1\int d\underline{r}_2\,\frac{e^{-\mu r_{12}}}{r_{12}}(1+\mu r_{12}-\mu^2 r_{12}^2)[1+f(\underline{r}_2)][1-f(\underline{r}_1)]P(\underline{r}_1,\underline{r}_2)$$

$$+\frac{k^2}{2N}\int d\underline{r}_1\int d\underline{r}_2\,\frac{e^{-\mu r_{12}}}{r_{12}}[\mu^2 r_{12}^2-\mu r_{12}-1][P(\underline{r}_1,\underline{r}_2)-\frac{N}{A}P(\underline{r}_1)-\frac{N}{A}P(\underline{r}_2)+\frac{N^2}{A^2}]$$

$$+\frac{k^2}{2N}\int d\underline{r}_1\int d\underline{r}_2\left[\frac{N}{A}P(\underline{r}_1)+\frac{N}{A}P(\underline{r}_2)-\frac{N^2}{A^2}\right]\frac{e^{-\mu r_{12}}}{r_{12}}\left[\mu^2 r_{12}^2-\mu r_{12}-1\right] \ . \tag{14}$$

The first term on the right hand side of Eq. (14) vanishes since we are now in the thermodynamic limit. The argument is identical to that used by Mermin[8]. The second term can also be seen to vanish after converting it to a surface integral

The next two terms also vanish but are trickier. Consider first the third term, here because of the integrand at least one of the integrations get restricted to Ω_a and let us call that integration r_2. Now

$$\frac{2}{N}\int_A d\underline{r}_1 \int_A d\underline{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}^3}(\mu^2 r_{12}^2 + \mu r_{12} + 1)[f(\underline{r}_1) - f(\underline{r}_2)]^2 P(\underline{r}_1, \underline{r}_2)$$

$$\le \frac{2}{N}\frac{b^2}{L}\int_A d\underline{r}_1 \int_{\Omega_a} d\underline{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}}(\mu^2 r_{12}^2 + \mu r_{12} + 1)\, P(\underline{r}_1, \underline{r}_2)$$

$$\sim \left(\frac{N}{A}\right) L^{-3/2} \tag{15}$$

Similarly the fourth term can be estimated to be

$$\frac{k^2}{2N}\int_A d\underline{r}_1 \int_A d\underline{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}}[1+f(\underline{r}_2)][1-f(\underline{r}_1)][1+\mu r_{12} - \mu^2 r_{12}^2] P(\underline{r}_1, \underline{r}_2)$$

$$\le \frac{5k^2}{4N}\int_{\Omega_a} d\underline{r}_1 \int d\underline{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}} P(\underline{r}_1, \underline{r}_2)$$

$$\sim \left(\frac{N}{A}\right) L^{-4} \tag{16}$$

In both of the estimates, Eqs. (15) and (16), we have followed the prescription that the thermodynamic limit is taken before $\mu \to 0$. If this were not the case both of these estimates would have diverged. Also, since we are in the thermodynamic limit the last term can also be seen to be zero by explicit integration. It is easy to verify that if we had made use of the function $\underline{B}$ as chosen by Mermin(2), the term analogous to the last term in Eq. (14) would have diverged as $\mu \to 0$. A closer look would reveal that the leading term in that case is proportional to k and not k^2, thus substantiating our earlier remarks about the choice of $\underline{B}$. We then have,

$$\lim_{k\to 0} \frac{4}{\pi}\int_0^{2\pi} d\theta_k\, \Lambda(\underline{k}) = \frac{k^2}{2N}\int d\underline{r}_1 \int d\underline{r}_2 \frac{e^{-\mu r_{12}}}{r_{12}}\left[P(\underline{r}_1, \underline{r}_2) - \frac{N}{A}P(\underline{r}_1) - \frac{N}{A}P(\underline{r}_2) + \frac{N^2}{A^2}\right](\mu^2 r_{12}^2 - \mu r_{12} - 1) \tag{17}$$

The limit $\mu \to 0$ now allows us to identify the right hand side of Eq. (17) as proportional to the internal energy per electron of a system of electrons embedded in a neutralizing uniform positive background and interacting by a potential e^2/r in two dimensions. Hence,

$$\lim_{\mu\to 0}\lim_{k\to 0} \frac{4}{\pi}\int_0^{2\pi} d\theta_k\, \Lambda(\underline{k}) = -k^2 (E_{int}/N) \tag{18}$$

For this system a rigorous lower bound has recently been obtained by Totsuji(6)

$$\frac{E_{int}}{N} \ge -\frac{1.1284\, e^2}{a} \tag{19}$$

where a is the average interparticle distance. Thus we have,

$$\lim_{\mu\to 0}\lim_{k\to 0} \frac{4}{\pi}\int_0^{2\pi} d\theta_k\, \Lambda(\underline{k}) \le k^2\left(\frac{1.1284\, e^2}{a}\right) \tag{20}$$

which can be used in the denominator to strengthen the inequality(13) further. One additional information is required. This is

$$\left|\frac{1}{N}\int d\underline{r}_1 P(\underline{r}_1) f(\underline{r}_1)\, e^{i\underline{Q}\cdot\underline{r}_1}\right|^2 \ge \left|\frac{\langle \mathcal{O}_Q \rangle}{N} - \frac{1}{N}\int_{\Omega_a} d\underline{r}_1 P(\underline{r}_1)\right|^2 \tag{21}$$

The right hand side of Eq. (21) can be replaced in the thermodynamic limit by $(\langle \mathcal{O}_Q \rangle / N)^2$ since by assumption the equilibrium state does not have macroscopic surface charge(7).

To complete the proof it needs to be shown that the left hand side of the inequality, Eq. (13), is finite. This will then imply that a finite quantity is greater than or equal to infinity and hence the inescapable conclusion that

The next two terms also vanish but are trickier. Consider first the third term, here because of the integrand at least one of the integrations get restricted to Ω_a and let us call that integration r_2. Now

$$\frac{2}{N}\int_A d\underline{r}_1\int_A d\underline{r}_2\ \frac{e^{-\mu r_{12}}}{r_{12}^3}\ (\mu^2 r_{12}^2+\mu r_{12}+1)[f(\underline{r}_1)-f(\underline{r}_2)]^2 P(\underline{r}_1,\underline{r}_2)$$

$$\le\ \frac{2}{N}\ \frac{b^2}{L}\ \int_A d\underline{r}_1\int_{\Omega_a} d\underline{r}_2\ \frac{e^{-\mu r_{12}}}{r_{12}}\ (\mu^2 r_{12}^2+\mu r_{12}+1)\ P(\underline{r}_1,\underline{r}_2)$$

$$\sim \left(\frac{N}{A}\right)L^{-\beta/2}$$

(15)

Similarly the fourth term can be estimated to be

$$\frac{k^2}{2N}\ \int_A d\underline{r}_1\int_A d\underline{r}_2\ \frac{e^{-\mu r_{12}}}{r_{12}}\ [1+f(\underline{r}_2)][1-f(\underline{r}_1)][1+\mu r_{12}-\mu^2 r_{12}^2]P(\underline{r}_1,\underline{r}_2)$$

$$\le\ \frac{5k^2}{4N}\ \int_{\Omega_a} dr_1\int dr_2\ \frac{e^{-\mu r_{12}}}{r_{12}}\ P(r_1,r_2)$$

$$\sim \left(\frac{N}{A}\right)L^{-\frac{1}{2}}$$

(16)

In both of the estimates, Eqs. (15) and (16), we have followed the prescription that the thermodynamic limit is taken before $\mu \to 0$. If this were not the case both of these estimates would have diverged. Also, since we are in the thermodynamic limit the last term can also be seen to be zero by explicit integration. It is easy to verify that if we had made use of the function $\underline{B}$ as chosen by Merein[2], the term analogous to the last term in Eq. (14) would have diverged as $\mu \to 0$. A closer look would reveal that the leading term in that case is proportional to k and not k^2, thus substantiating our earlier remarks about the choice of $\underline{B}$. We then have,

$$\lim_{k\to 0}\ \frac{4}{\pi}\int_0^{2\pi} d\theta_k\, \Lambda(\underline{k}) = \frac{k^2}{2N}\int d\underline{r}_1\int d\underline{r}_2\ \frac{e^{-\mu r_{12}}}{r_{12}}\Big[P(\underline{r}_1,\underline{r}_2)-\frac{N}{A}P(\underline{r}_1)-\frac{N}{A}P(\underline{r}_2)+\frac{N^2}{A^2}\ldots r_{12}^2 \ldots r_{12}-1\Big]$$

(17)

The limit $\mu \to 0$ now allows us to identify the right hand side of Eq. (17) as proportional to the internal energy per electron of a system of electrons embedded in a neutralizing uniform positive background and interacting by a potential e^2/r in two dimensions. Hence,

$$\lim_{\mu\to 0}\ \lim_{k\to 0}\ \frac{4}{\pi}\int_0^{2\pi} d\theta_k\, \Lambda(\underline{k}) = -k^2(E_{int}/N)$$

(18)

For this system a rigorous lower bound has recently been obtained by Totsuji[8].

$$\frac{E_{int}}{N} \ge -\frac{1.1284\ e^2}{a}$$

(19)

where a is the average interparticle distance. Thus we have,

$$\lim_{\mu\to 0}\ \lim_{k\to 0}\ \frac{4}{\pi}\int_0^{2\pi} d\theta_k\, \Lambda(\underline{k}) \le k^2\left(\frac{1.1284\ e^2}{a}\right)$$

(20)

which can be used in the denominator to strengthen the inequality[13] further. One additional information is required. This is

$$\left|\frac{1}{N}\int d\underline{r}_1 P(\underline{r}_1) f(\underline{r}_1)\ e^{i\underline{k}\cdot\underline{r}_1}\right|^2 \ge \left|\frac{\langle \mathcal{C}_c\rangle}{N}-\frac{1}{N}\int_{\Omega_a} d\underline{r}_1 P(\underline{r}_1)\right|^2$$

(21)

The right hand side of Eq. (21) can be replaced in the thermodynamic limit by $(\langle \mathcal{C}_c\rangle/N)^2$ since by assumption the equilibrium state does not have macroscopic surface charge[7].

To complete the proof it needs to be shown that the left hand side of the inequality, Eq. (13), is finite. This will then imply that a finite quantity is greater than or equal to infinity and hence the inescapable conclusion that

$\langle \rho_c \rangle / N$ vanishes at any finite temperature. One way to proceed is to follow Mermin[2]. The important ingredient is of course the free energy of the system considered in this paper. This, thanks to Totsuji[9], can be shown to have both an upper and a lower bound. The other alternative is to follow Sorokina[10] word for word and arrive at the conclusion that the left hand side of the inequality (13) is bounded.

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